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(71)Applicant : JAPAN ENERGY CORP
(72)Inventor : HOSHI HIDEO

(54) MULTILAYER SEMICONDUCTOR SUBSTRATE AND PRODUCTION THEREOF

(57)Abstract:
PURPOSE: To obtain a multilayer semiconductor substrate comprising a thin metal oxide deposited uniformly on a silicon semiconductor substrate and a production method thereof.
CONSTITUTION: A semiconductor substrate and a material metal are set in an epitaxial growth system and a metal film is grown, at first, on the semiconductor substrate at a temperature of ≤ 250° C. Subsequently, oxygen gas is introduced into the epitaxial growth system in order to deposit a metal oxide on the metal film in the oxygen atmosphere. With such method, a thin metal oxide is deposited uniformly on the semiconductor substrate and an excellent substrate suitable for fabricating a nonvoltile memory, a capacitor, a thin film superconducting device, an optical conversion element, etc., is obtained.

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(71)出願人　000231109
株式会社ジャパンエナジー
東京都港区虎ノ門二丁目10番１号

(72)発明者　星　英夫
埼玉県戸田市新曽南３丁目17番35号　株式会社ジャパンエナジー内

(74)代理人　弁理士　荒船　博司

(54)【発明の名称】　多層半導体基板及びその製造方法

(57)【要約】

【目的】　シリコン半導体基板上に薄くて均一な希土類元素の金属酸化物膜を有する多層半導体基板及びその製造方法を提供する。

【構成】　エピタキシャル成長装置内に、半導体基板と金属原料を入れ、まず２５０℃以下の成長温度で該半導体基板上に金属膜を成長させ、その後、該エピタキシャル成長装置内に酸素ガスを導入し、酸素雰囲気中で前記金属膜上に金属酸化物膜を形成する。

【効果】　半導体基板上に薄くて均一な金属酸化物膜が形成され、不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などの作製用基板として好適な優れた基板が得られる。

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【特許請求の範囲】

【請求項１】　半導体基板上に不活性ガス成分を含まないエピタキシャル成長された金属酸化物膜が形成され、かつその界面には前記金属酸化物の勾配層の厚さが５原子層以下に相当する厚さの酸素の組成勾配層が形成されていることを特徴とする多層半導体基板。

【請求項２】　前記半導体基板は、面方位が（１１１）または（１００）の単結晶シリコンでできていることを特徴とする請求項１記載の多層半導体基板。

【請求項３】　前記金属酸化物膜は、Ｃ希土構造を有する酸化物であることを特徴とする請求項１または２記載の多層半導体基板。

【請求項４】　前記金属酸化物膜は、酸化イットリウムまたは酸化ジスプロシウムまたはイットリア部分安定化ジルコニアでできていることを特徴とする請求項１または２記載の多層半導体基板。

【請求項５】　エピタキシャル成長装置内に、半導体基板と金属原料を入れ、まず２５０℃以下の成長温度で該半導体基板上に金属膜を成長させ、続いて該エピタキシャル成長装置内に酸素ガスを導入し、酸素雰囲気中で前記金属膜上に金属酸化物膜を形成することを特徴とする多層半導体基板の製造方法。

【請求項６】　前記半導体基板として、面方位が（１１１）または（１００）の単結晶シリコンよりなる基板を用いることを特徴とする請求項５記載の多層半導体基板の製造方法。

【請求項７】　前記金属原料として、酸化したときにＣ希土構造となる金属を用い、この金属よりなる前記金属膜を成長させるとともに、Ｃ希土構造の金属酸化物よりなる前記金属酸化物膜を成長させることを特徴とする請求項５または６記載の多層半導体基板の製造方法。

【請求項８】　前記金属原料として、イットリウムまたはジスプロシウムを用い、イットリウムまたはジスプロシウムよりなる前記金属膜を成長させるとともに、酸化イットリウムまたは酸化ジスプロシウムよりなる前記金属酸化物膜を成長させることを特徴とする請求項５または６記載の多層半導体基板の製造方法。

【請求項９】　前記金属原料として、ジルコニウムを用い、ジルコニウムよりなる前記金属膜を成長させるとともに、イットリア安定化ジルコニアよりなる前記金属酸化物膜を成長させることを特徴とする請求項５または６記載の多層半導体基板の製造方法。

【請求項１０】　前記金属膜の成長を、１原子層以上５原子層以下に相当する厚さになるまで行うことを特徴とする請求項５、６、７、８または９記載の多層半導体基板の製造方法。

【請求項１１】　前記金属膜の成長を、１．５原子層以上２．５原子層以下に相当する厚さになるまで行うことを特徴とする請求項５、６、７、８または９記載の多層半導体基板の製造方法。

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【請求項１２】　前記半導体基板の表面をあらかじめ水素終端処理しておくことを特徴とする請求項６記載の多層半導体基板の製造方法。

【発明の詳細な説明】

【０００１】

【産業上の利用分野】本発明は、多層構造の半導体基板に関し、例えば高機能の不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などの素子作製用の基板及びその基板の製造方法に関する。

【０００２】

【従来の技術】高機能の不揮発性メモリーやキャパシター、或いは光変調素子などを作製する場合に使用可能な配向性強誘電体薄膜素子として、特開平６－１９６６４８号公報に記載されているように、ＧａＡｓ単結晶基板上に、金属酸化物膜よりなるバッファ薄膜、金属薄膜及び強誘電体薄膜を順次積層した構造のものが知られている。

【０００３】このような構造の素子を単結晶シリコンよりなる半導体基板上に作製する場合には、まず半導体基板上に金属酸化物膜を蒸着法やスパッタ法などによりエピタキシャル成長させる。従来、その蒸発源やスパッタリングのターゲットには、金属酸化物の焼結体などが用いられている。しかしながら、上記蒸発源を電子ビーム等で加熱して蒸発させる際や、ターゲットをイオン衝撃する際に、酸化物からの酸素の解離が起こり、その解離した酸素によりシリコン半導体基板の表面が酸化されてしまい、特性の良い金属酸化物膜が得られないという問題点があった。

【０００４】この問題に対し、表面に酸化膜を有するシリコン半導体基板上に、８０Åの金属ストロンチウムを成膜した後チタン酸ストロンチウムエピタキシャル膜を成膜することで、金属ストロンチウムによりシリコン半導体基板表面の酸化膜を還元させ良好なバッファ膜が得られることが開示されている（第１３回マテリアルサイエンスアンドエンジニアリングシンポジウム論文集（Proceedings of the 13th Symposium on Material Science and Engineering）１０９頁～１１４頁（１９９４））。しかし、この方法では、金属ストロンチウムの膜厚が８０Åと厚いため、チタン酸ストロンチウム膜の組成ずれが生じてしまう。また、チタン酸ストロンチウム膜の膜厚は約１０００Åと推測される（論文中には明記されていない）が、バッファ膜としては厚いものである。

【０００５】また、シリコン半導体基板上に、スパッタリング法により室温で４ÅのＺｒ$_{1-x}$ Ｙ$_{x}$ を成膜し、その上に２４００Åのイットリア部分安定化ジルコニアエピタキシャル膜を成膜する技術が開示されている（シンソリッドフィルム（Thin Solid Films）２２９巻、１７頁～２３頁（１９９３））。しかし、この方法は、スパッタリング法を用いているため、膜中の不活性ガス濃度

が数％と高くなってしまい、この上に更に強誘電体膜、超電導体膜等の成膜を行い素子化したときの電気的特性、信頼性の上で問題が生じる。また、この技術も、イットリア部分安定化ジルコニア膜の膜厚は２４００Åとバッファ膜としては厚いものである。

【０００６】

【発明が解決しようとする課題】本発明は、上記問題点を解決するためになされたもので、その目的は、半導体基板上に薄くて均一な金属酸化物膜を有し、高機能の不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などの素子作製用の基板として好適な多層半導体基板を提供することにある。

【０００７】また、本発明の他の目的は、半導体基板上に薄くて均一な金属酸化物膜を形成することのできる多層半導体基板の製造方法を提供することである。

【０００８】

【課題を解決するための手段】上記目的を達成するために、本発明者は、“シリコン系ヘテロデバイス”（古川ら、丸善、平成３年７月３０日発行）の１５０頁に記載された「O_2 ガスとシリコン表面の反応」の図（同書図７．４、同図に対応する領域を本明細書に添付した図面の図１の右上に一点鎖線で区切って示す。）に基づいて、さらに研究を行った。その結果、図１に示すように、O_2 ガス圧力が10^{-10} Paで基板温度が室温から成長温度（８００℃）に達するまでに、略２６０～５００℃の領域（酸化領域II）において基板表面が酸化され、続く略５００℃以上の領域（エッチング領域III ）で基板表面がエッチングされることがわかった。なお、実際の金属酸化物膜の成長では、温度及びO_2 ガスの圧力を、図１中のイーロ－ハ線に沿ってイ点からハ点に向かって変化させている。

【０００９】そこで、本発明者は、基板温度の昇温過程において、略２６０～５００℃の酸化領域IIに至る前、すなわち例えば２００～２５０℃程度の基板温度でもって、基板表面を金属薄膜で被覆することによりその酸化領域IIにおける基板表面の酸化を防止することができると考えた。

【００１０】本発明は上記知見等に基づいてなされたもので、請求項１記載の多層半導体基板は、半導体基板上に不活性ガス成分を含まないエピタキシャル成長された金属酸化物膜が形成され、かつその界面には前記金属酸化物の勾配層の厚さが５原子層以下に相当する厚さの酸素の組成勾配層が形成されていることを特徴とする。

【００１１】この発明において、請求項２記載の発明のように、前記半導体基板は、面方位が（１１１）または（１００）の単結晶シリコンでできていてもよいし、請求項３または４記載の発明のように、前記金属酸化物膜は、Ｃ希土構造を有する酸化物、酸化イットリウム、酸化ジスプロシウム、イットリア部分安定化ジルコニアであってもよい。

【００１２】上記多層半導体基板を製造するにあたって、請求項５記載の発明は、エピタキシャル成長装置内に、半導体基板と金属原料を入れ、まず２５０℃以下の成長温度で該半導体基板上に金属膜を成長させ、続いて該エピタキシャル成長装置内に酸素ガスを導入し、酸素雰囲気中で前記金属膜上に金属酸化物膜を形成することを特徴とする。

【００１３】この発明において、請求項６記載の発明のように、前記半導体基板として、面方位が（１１１）または（１００）の単結晶シリコンよりなる基板を用いてもよいし、請求項７記載の発明のように、前記金属原料として、酸化したときにＣ希土構造となる金属を用い、この金属よりなる前記金属膜を成長させるとともに、Ｃ希土構造の金属酸化物よりなる前記金属酸化物膜を成長させるようにしてもよい。また、請求項８記載の発明のように、前記金属原料として、イットリウムまたはジスプロシウムを用い、イットリウムまたはジスプロシウムよりなる前記金属膜を成長させるとともに、酸化イットリウムまたは酸化ジスプロシウムよりなる前記金属酸化物膜を成長させてもよいし、請求項９記載の発明のように、前記金属原料として、ジルコニウムを用い、ジルコニウムよりなる前記金属膜を成長させるとともに、イットリア安定化ジルコニアよりなる前記金属酸化物膜を成長させてもよい。そして、請求項１０記載の発明のように、前記金属膜の成長を、１原子層以上５原子層以下に相当する厚さになるまで行うようにしてもよいし、或いは請求項１１記載の発明のように、前記金属膜の成長を、１．５原子層以上２．５原子層以下に相当する厚さになるまで行うようにしてもよい。

【００１４】また、請求項１２記載の発明のように、前記半導体基板の表面をあらかじめ水素終端処理しておいてもよい。

【００１５】

【作用】本発明に係る多層半導体基板によれば、半導体基板上に不活性ガス成分を含まないエピタキシャル成長された金属酸化物膜が形成され、かつその界面には勾配層の厚さが５原子層以下に相当する厚さの前記金属酸化物の酸素の組成勾配層が形成された、薄くて均一な金属酸化物膜を有する多層半導体基板である。その製造プロセスにおいて、まず２５０℃以下の成長温度で半導体基板上に金属膜を成長させ、続いてエピタキシャル成長装置内に酸素ガスを導入し、酸素雰囲気中で前記金属膜上に金属酸化物膜を成長させる。このとき５原子層より金属膜が厚いと、金属膜成膜後の昇温過程において半導体基板中に金属が拡散しやすくなる。また、昇温後の金属酸化物膜の成膜中に酸素がこの金属膜中に拡散するので、半導体基板と金属酸化物の界面には、５原子層以下に相当する厚さに相当するこの金属膜と酸素の反応した金属酸化物膜の酸素の組成勾配層が形成される。従って、この基板を用いて不揮発性メモリーやキャパシタ

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一、薄膜超電導デバイス、或いは光変調素子などの素子を作製すると、金属酸化物膜よりなるバッファ層が薄く、優れた特性の素子が得られる。

【００１６】そして、半導体基板が面方位が（１１１）または（１００）の単結晶シリコンでできていることにより、不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などとを集積化することができる。また、金属酸化物膜がＣ希土構造を有する酸化物、酸化イットリウム、酸化ジスプロシウムまたはイットリア部分安定化ジルコニアでできていれば、得られた多層半導体基板は、不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などの素子作製用の基板として好適である。なお、Ｃ希土構造を有する酸化物としては、酸化イットリウム（Y_2O_3）、酸化ジスプロシウム（Dy_2O_3）の他に、酸化ネオジウム（Nd_2O_3）、酸化サマリウム（Sm_2O_3）、酸化ユーロピウム（Eu_2O_3）、酸化プロシチウム（Pr_2O_3）等がある。

【００１７】本発明に係る多層半導体基板の製造方法によれば、エピタキシャル成長装置内に、半導体基板と金属原料を入れ、まず２５０℃以下の成長温度で該半導体基板上に金属膜を成長させ、続いて該エピタキシャル成長装置内に酸素ガスを導入し、酸素雰囲気中で前記金属膜上に金属酸化物膜を成長させるようにしたため、金属酸化物の成長温度（例えば８００℃）まで基板温度を昇温させる過程において、半導体基板の表面が酸化及びエッチングされるのが防止されるので、薄くて均一な金属酸化物薄膜が形成される。従って、不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などの作製用基板として好適な優れた基板が得られる。

【００１８】そして、面方位が（１１１）または（１００）の単結晶シリコン半導体基板を用いることにより、不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などの半導体素子と集積化させるのに好適な基板が得られる。また、金属膜として酸化したときにＣ希土構造となる金属よりなる前記金属膜を成長させるとともに、金属酸化物膜としてＣ希土構造の金属酸化物よりなる膜を成長させる、または金属膜としてイットリウムまたはジスプロシウムよりなる前記金属膜を成長させるとともに、金属酸化物膜として酸化イットリウムまたは酸化ジスプロシウムよりなる膜を成長させる、或いは金属膜としてジルコニウムよりなる前記金属膜を成長させるとともに、金属酸化物膜としてイットリア安定化ジルコニアよりなる膜を成長させることにより、優れた多層半導体基板が得られる。さらに、金属膜を、その厚さが１～５原子層、好ましくは１．５～２．５原子層以下に相当する厚さとなるように成長させることにより、薄くて均一な金属酸化物薄膜を有し、かつ半導体基板が金属膜中の金属原子により汚染されていない多層基板が得られる。金属膜の厚さが１原子層未満と薄

6

すぎると、半導体基板表面の酸化防止やエッチング防止の効果が十分に得られない。逆に、金属膜の厚さが５原子層より厚すぎると、金属膜成膜後、基板温度を上げていったときに金属膜から半導体基板中への金属原子が拡散しやすくなる。

【００１９】また、半導体基板の表面をあらかじめ希フッ酸等の希酸溶液やフッカアンモニウム水に曝したり、半導体基板の表面に水素ラジカルを照射したりして、基板表面に水素終端処理しておくことにより、半導体基板の酸化防止の効果が高くなり、清浄な半導体基板表面に１～５原子層程度の厚さの薄くて均一な金属酸化物膜を有する多層半導体基板が得られる。

【００２０】

【実施例】以下に、実施例を挙げて本発明の特徴とするところを明らかとする。なお、本発明は以下の各実施例により何等制限されるものではないのはいうまでもない。

【００２１】（実施例１）まず、周知の蒸着装置を用い、成長室内を真空排気して真空雰囲気（圧力：1×10^{-6}Pa）とした後、面方位が（１１１）の単結晶シリコンよりなる半導体基板上にＹ（イットリウム）膜を成長させた。原料には、Ｙの単体（金属）を用いた。基板の温度は２５０℃であった。成長したＹ膜の厚さは４Åであった。なお、半導体基板は、あらかじめ希フッ酸（dil HF）等の酸でウェット処理を行い、半導体基板の表面の水素終端処理をしておいたものを使用した。

【００２２】続いて、成長室内にO_2 ガスを導入し、酸素雰囲気（圧力：1×10^{-2}Pa）中で、先に成長させたＹ膜上にY_2O_3 膜をエピタキシャル成長させた。原料には、Ｙの単体を用いた。基板温度は７００℃であった。成長したY_2O_3 膜の厚さは２００Åであった。

【００２３】図２（ａ）にＸＲＤ（Ｘ線回折）及び図２（ｂ）にＲＨＥＥＤ（反射高速電子線回折）の結果を模式的に示した図を示す。図２（ａ）（ｂ）からわかるように、ＸＲＤの結果には、Y_2O_3（４４４）のピークとＳｉ（１１１）基板のピークだけが現れており、また、ＲＨＥＥＤの結果には、明確なパターンが現れている。これらの結果より、Ｓｉ（１１１）基板上に表面に荒れの少ない均一なY_2O_3（１１１）配向のエピタキシャル膜が成長しているのがわかる。

【００２４】（実施例２）上記実施例１と同様にして、まず、真空雰囲気（圧力：1×10^{-6}Pa）中で、面方位が（１１１）の単結晶シリコンよりなる半導体基板（あらかじめその表面を酸でウェット処理を行い、半導体基板の表面の水素終端処理をしておいた。）上にＤｙ（ジスプロシウム）膜を成長させた。原料には、Ｄｙの単体（金属）を用いた。基板の温度は２５０℃であった。成長したＤｙ膜の厚さは４Åであった。

【００２５】続いて、酸素雰囲気（圧力：1×10^{-2}Pa）中で、Ｄｙ膜上にDy_2O_3 膜をエピタキシャル成

長させた。原料には、Ｄｙの単体を用いた。基板温度は７００℃であった。成長したDy_2O_3膜の厚さは２００Åであった。

【００２６】図３（ａ）にＸＲＤ及び図３（ｂ）にＲＨＥＥＤの結果を模式的に示した図を示す。図３（ａ）（ｂ）からわかるように、ＸＲＤの結果には、Dy_2O_3（４４４）のピークとＳｉ（１１１）基板のピークだけが現れており、また、ＲＨＥＥＤの結果には、明確なパターンが現れている。これらの結果より、Ｓｉ（１１１）基板上に表面に荒れの少ない均一なDy_2O_3（１１１）配向のエピタキシャル膜が成長しているのがわかる。

【００２７】（実施例３）上記実施例１と同様にして、まず、真空雰囲気（圧力：1×10^{-6}Pa）中で、面方位が（１００）の単結晶シリコンよりなる半導体基板（あらかじめその表面を酸でウェット処理を行い、半導体基板の表面の水素終端処理をしておいた。）上にＺｒ膜を成長させた。原料には、Ｚｒの単体（金属）を用いた。基板の温度は２５０℃であった。成長したＺｒ膜の厚さは４Åであった。

【００２８】続いて、成長室内にO_2ガスを導入し、酸素雰囲気（圧力：5×10^{-3}Pa）中で、先に成長させたＺｒ膜上にＹＳＺ（イットリア安定化ジルコニア（組成；Y_2O_3：ZrO_2＝９：９１（モル比））膜を成長させた。原料には、ＹＳＺの単体を用いた。基板温度は８００℃であった。成長したＹＳＺ膜の厚さは２００Åであった。

【００２９】図４（ａ）にＸＲＤ及び図４（ｂ）にＲＨＥＥＤの結果を模式的に示した図を示す。図４（ａ）（ｂ）からわかるように、ＸＲＤの結果には、ＹＳＺ（２００）のピークだけが現れており、また、ＲＨＥＥＤの結果には、明確なパターンが現れている（なお、ＸＲＤの結果（図４（ａ））にはＳｉ（１００）基板のピークは現れていない）。これらの結果より、Ｓｉ（１００）基板上に表面に荒れの少ない均一なＹＳＺ（１００）配向のエピタキシャル膜が成長しているのがわかる。

【００３０】以上のように、上記実施例１～３で得られた各試料（多層半導体基板）についていずれの試料もその表面に荒れの少ない均一なエピタキシャル膜が成長しているのがわかる。

【００３１】上記実施例によれば、半導体基板上に真空雰囲気中で金属薄膜を成長させ、さらにその上に酸素雰囲気中で金属酸化物薄膜を成長させることにより、半導体基板上に薄くて均一な金属酸化物薄膜よりなるバッファー層を形成することができ、不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などを作製する際に使用する基板として優れた特性を有する多層半導体基板が得られる。

【００３２】なお、蒸発源（原料）として用いる金属は、上記実施例に限らない。

【００３３】また、半導体基板は、シリコン基板に限らないのは勿論である。

【００３４】

【発明の効果】本発明に係る多層半導体基板によれば、半導体基板上に薄くて均一な金属酸化物膜を有する多層半導体基板が得られるので、この基板を用いることにより、特性の優れた不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などの素子が得られる。また、その様な各種素子と半導体素子とを集積化することができる。

【００３５】本発明に係る多層半導体基板の製造方法によれば、半導体基板上に薄くて均一な金属酸化物膜が形成され、不揮発性メモリーやキャパシター、薄膜超電導デバイス、或いは光変調素子などの作製用基板として好適な優れた基板が得られる。

【図面の簡単な説明】

【図１】O_2ガスとシリコン表面の反応の特性を示す図である。

【図２】実施例１の結果を示した図であり、図２（ａ）はＸＲＤの結果を示す図、図２（ｂ）はＲＨＥＥＤの結果を模式的に示す図である。

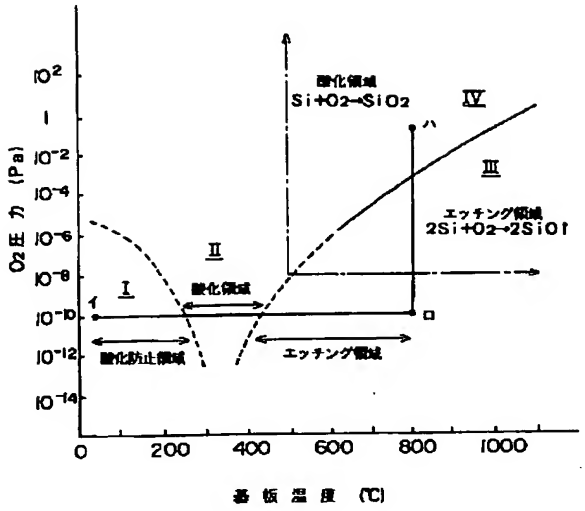
【図３】実施例２の結果を示した図であり、図３（ａ）はＸＲＤの結果を示す図、図３（ｂ）はＲＨＥＥＤの結果を模式的に示す図である。

【図４】実施例３の結果を示した図であり、図４（ａ）はＸＲＤの結果を示す図、図４（ｂ）はＲＨＥＥＤの結果を模式的に示す図である。

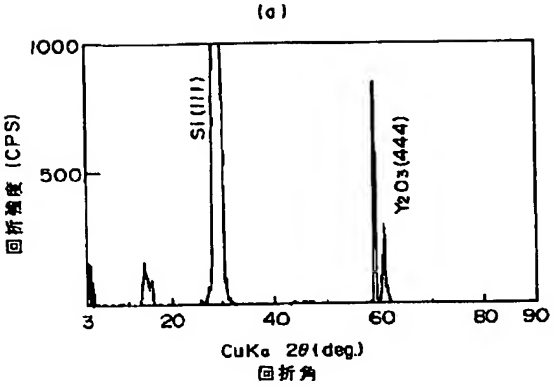
【符号の説明】

I　酸化防止領域
II　酸化領域
III　エッチング領域
IV　酸化領域

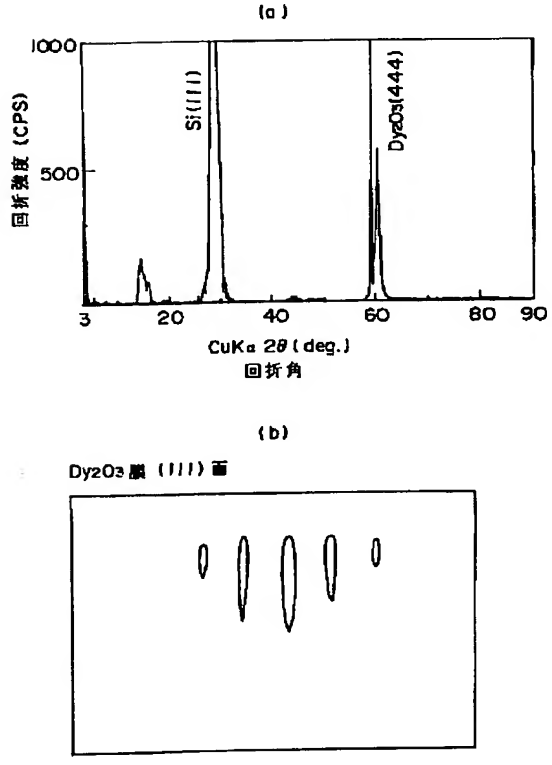
【図１】


酸化領域
Si+O2→SiO2
エッチング領域
2Si+O2→2SiOf
I
II
III
IV
酸化防止領域
エッチング領域
O2圧力 (Pa)
基板温度 (℃)

【図２】

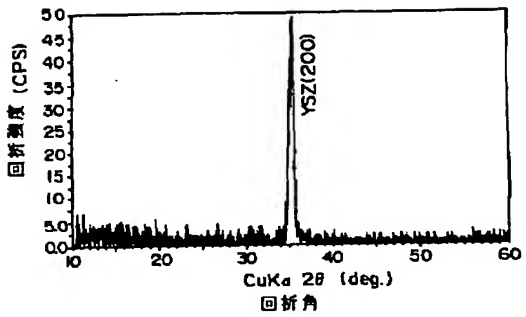

(a)
Si(111)
Y2O3(444)
回折強度 (CPS)
CuKα 2θ(deg.)
回折角
(b)
Y2O3膜 (111)面

【図３】

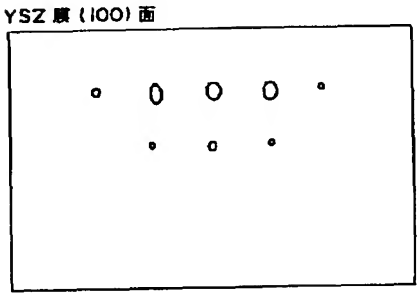

(a)
Si(111)
Dy2O3(444)
回折強度 (CPS)
CuKα 2θ (deg.)
回折角
(b)
Dy2O3膜 (111)面

【図４】

(a)



(b)



フロントページの続き

(51) Int. Cl.[6]	識別記号	庁内整理番号	Ｆ Ｉ	技術表示箇所
Ｈ０１Ｌ　21/8242				

* NOTICES

1. This document has been translated by computer.So the translation may not reflect the original precisely.
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CLAIMS

[Claim(s)]
[Claim 1] The multilayer semiconductor substrate characterized by forming the epitaxially grown metallic-oxide film which does not contain an inert gas component on a semiconductor substrate, and forming in the interface the composition inclination layer of the oxygen of the thickness by which the inclination layer thickness of the aforementioned metallic oxide is equivalent to below a pentatomic layer.
[Claim 2] the aforementioned semiconductor substrate -- a field direction -- or (111) (100) the multilayer semiconductor substrate according to claim 1 characterized by being made of single crystal silicon
[Claim 3] The aforementioned metallic-oxide film is a multilayer semiconductor substrate according to claim 1 or 2 characterized by being the oxide which has C rare-earth structure.
[Claim 4] The aforementioned metallic-oxide film is a multilayer semiconductor substrate according to claim 1 or 2 characterized by being made of a yttrium oxide, an oxidization dysprosium, or a yttria partially stabilized zirconia.
[Claim 5] The manufacture method of the multilayer semiconductor substrate characterized by paying a semiconductor substrate and a metal raw material in an epitaxial growth system, growing up a metal membrane on this semiconductor substrate, introducing oxygen gas in this epitaxial growth system continuously at the growth temperature of 250 degrees C or less first, and forming a metallic-oxide film on the aforementioned metal membrane in oxygen atmosphere.
[Claim 6] as the aforementioned semiconductor substrate -- a field direction -- or (111) (100) the manufacture method of the multilayer semiconductor substrate according to claim 5 characterized by using the substrate which consists of single crystal silicon
[Claim 7] The manufacture method of the multilayer semiconductor substrate according to claim 5 or 6 characterized by growing up the aforementioned metallic-oxide film which consists of a metallic oxide of C rare-earth structure while growing up the aforementioned metal membrane which consists of this metal as the aforementioned metal raw material using the metal used as C rare-earth structure, when it oxidizes.
[Claim 8] The manufacture method of the multilayer semiconductor substrate according to claim 5 or 6 characterized by growing up the aforementioned metallic-oxide film which consists of a yttrium oxide or an oxidization dysprosium while growing up the aforementioned metal membrane which consists of an yttrium or a dysprosium, using an yttrium or a dysprosium as the aforementioned metal raw material.
[Claim 9] The manufacture method of the multilayer semiconductor substrate according to claim 5 or 6 characterized by growing up the aforementioned metallic-oxide film which consists of a yttria stabilized zirconia while growing up the aforementioned metal membrane which consists of a zirconium, using a zirconium as the aforementioned metal raw material.
[Claim 10] The manufacture method of the multilayer semiconductor substrate according to claim 5, 6, 7, 8, or 9 characterized by performing growth of the aforementioned metal membrane until it becomes the thickness equivalent to five or less atomic layer more than per atomic layer.
[Claim 11] The manufacture method of the multilayer semiconductor substrate according to claim 5, 6,

7, 8, or 9 characterized by performing growth of the aforementioned metal membrane until it becomes the thickness equivalent to 1.5 or more-atomic layer 2.5 or less atomic layer.
[Claim 12] The manufacture method of the multilayer semiconductor substrate according to claim 6 characterized by carrying out hydrogen termination processing of the front face of the aforementioned semiconductor substrate beforehand.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Industrial Application] this invention relates to the substrate for element production of a highly efficient nonvolatile memory and a highly efficient capacitor, a thin film superconductivity device, or an optical modulator, and the manufacture method of the substrate, concerning the semiconductor substrate of multilayer structure.
[0002]
[Description of the Prior Art] When producing a highly efficient nonvolatile memory, a capacitor or an optical modulator, etc., the thing of the structure which carried out the laminating of the buffer thin film, metal thin film, and ferroelectric thin film which consist of a metallic-oxide film on a GaAs single crystal substrate one by one is known as an usable stacking-tendency ferroelectric thin film as indicated by JP,6-196648,A.
[0003] In producing the element of such structure on the semiconductor substrate which consists of single crystal silicon, it grows a metallic-oxide film epitaxially by the vacuum deposition, the spatter, etc. on a semiconductor substrate first. Conventionally, the sintered compact of a metallic oxide etc. is used for the target of the evaporation source and sputtering. However, when heating the above-mentioned evaporation source and evaporating it with an electron beam etc., or when carrying out the ion bombardment of the target, the maceration of the oxygen from an oxide happened, the front face of a silicon semiconductor substrate will oxidize by the dissociated oxygen, and there was a trouble that a metallic-oxide film with a sufficient property was not obtained.
[0004] By forming a strontium-titanate epitaxial film, after forming 80A metal strontium to this problem on the silicon semiconductor substrate which has an oxide film on a front face With metal strontium The oxide film of a silicon semiconductor substrate front face It is indicated that make it return and a good buffer film is obtained (the 13th material science and engineering symposium collected works ()).
[Proceedings of the 13 th Symposium on Material] 109 pages - 114 page (1994) of Science and Engineering. However, by this method, since the thickness of metal strontium is as thick as 80A, a composition gap of a strontium-titanate film will arise. moreover, the thickness of a strontium-titanate film conjectures about 1000A -- having (not clearly written in the paper) -- as a buffer film, it is thick
[0005] Moreover, it is 4A Zr1-x Yx at a room temperature by the sputtering method on a silicon semiconductor substrate. The technology which forms membranes and forms a 2400A yttria partially-stabilized-zirconia epitaxial film on it is indicated (229 Thin Solid Films (Thin Solid Films), 17 pages - 23 pages (1993)). However, since the sputtering method is used for this method, the inert gas concentration in a film becomes high with several %, and a problem produces it on the electrical property when forming a ferroelectric film, a superconductor film, etc. further and element-izing on this, and reliability. Moreover, this technology of the thickness of a yttria partially-stabilized-zirconia film is also thick as 2400A and a buffer film.
[0006]
[Problem(s) to be Solved by the Invention] this invention was made in order to solve the above-

mentioned trouble, it has a metallic-oxide film thin [on a semiconductor substrate] the purpose and uniform, and is to offer a multilayer semiconductor substrate suitable as a substrate for element production of a highly efficient nonvolatile memory and a highly efficient capacitor, a thin film superconductivity device, or an optical modulator.
[0007] Moreover, other purposes of this invention are offering the manufacture method of the multilayer semiconductor substrate which can form a thin and uniform metallic-oxide film on a semiconductor substrate.
[0008]
[Means for Solving the Problem] in order to attain the above-mentioned purpose -- this invention person -- "a silicon system hetero device" (Furukawa et al. --) Maruzen, drawing of "the reaction on O2 gas and the front face of silicon" indicated by 150 pages of issue on July 30, Heisei 3 (an alternate long and short dash line divides and shows the field corresponding to this writing view 7.4 and this drawing to the upper right of drawing 1 of the drawing appended to this specification.) It was based and inquired further. Consequently, it is O2 as shown in drawing 1 . It turns out that a substrate front face will oxidize in the field (oxidization field II) of 260-500 degrees C of abbreviation by the time substrate temperature reaches [gas pressure] growth temperature (800 degrees C) from a room temperature in 10-10 Pa, and a substrate front face **********s in the field (etching field III) of 500 degrees C or more of continuing abbreviation. In addition, by growth of an actual metallic-oxide film, it is temperature and O2. The pressure of gas is changed from the I point toward a HA point along with the I-Law HA line in drawing 1 .
[0009] Then, in the temperature up process of substrate temperature, before this invention person reached the oxidization field II of 260-500 degrees C of abbreviation, he had at the substrate temperature of about 200-250 degrees C, and thought that oxidization on the front face of a substrate in the oxidization field II could be prevented by covering a substrate front face with a metal thin film.
[0010] A multilayer semiconductor substrate according to claim 1 is characterized by having not made this invention based on the above-mentioned knowledge etc., and forming the epitaxially grown metallic-oxide film which does not contain an inert gas component on a semiconductor substrate, and forming in the interface the composition inclination layer of the oxygen of the thickness by which the inclination layer thickness of the aforementioned metallic oxide is equivalent to below a pentatomic layer.
[0011] this invention -- setting -- invention according to claim 2 -- like -- the aforementioned semiconductor substrate -- a field direction -- or (111) (100) it may be made of single crystal silicon, and the aforementioned metallic-oxide films may be the oxide and yttrium oxide which have C rare-earth structure, an oxidization dysprosium, and a yttria partially stabilized zirconia like invention according to claim 3 or 4
[0012] In manufacturing the above-mentioned multilayer semiconductor substrate, invention according to claim 5 pays a semiconductor substrate and a metal raw material in an epitaxial growth system, first, grows up a metal membrane on this semiconductor substrate, introduces oxygen gas in this epitaxial growth system continuously at the growth temperature of 250 degrees C or less, and is characterized by forming a metallic-oxide film on the aforementioned metal membrane in oxygen atmosphere.
[0013] Or (111) (100) may use the substrate which consists of single crystal silicon. this invention -- setting -- invention according to claim 6 -- like -- as the aforementioned semiconductor substrate -- a field direction -- like invention according to claim 7 When it oxidizes, while growing up the aforementioned metal membrane which consists of this metal as the aforementioned metal raw material using the metal used as C rare-earth structure, you may make it grow up the aforementioned metallic-oxide film which consists of a metallic oxide of C rare-earth structure. Moreover, the aforementioned metallic-oxide film which consists of a yttrium oxide or an oxidization dysprosium while growing up the aforementioned metal membrane which consists of an yttrium or a dysprosium like invention according to claim 8, using an yttrium or a dysprosium as the aforementioned metal raw material may be grown up, and like invention according to claim 9, while growing up the aforementioned metal membrane which consists of a zirconium, using a zirconium as the aforementioned metal raw material, you may

grow up the aforementioned metallic-oxide film which consists of a yttria stabilized zirconia. And like invention according to claim 10, it may be made to perform growth of the aforementioned metal membrane until it becomes the thickness equivalent to five or less atomic layer more than per atomic layer, or like invention according to claim 11, you may be made to perform growth of the aforementioned metal membrane until it becomes the thickness equivalent to 1.5 or more-atomic layer 2.5 or less atomic layer.
[0014] Moreover, you may carry out hydrogen termination processing of the front face of the aforementioned semiconductor substrate beforehand like invention according to claim 12.
[0015]
[Function] According to the multilayer semiconductor substrate concerning this invention, it is the multilayer semiconductor substrate which has the thin and uniform metallic-oxide film with which the epitaxially grown metallic-oxide film which does not contain an inert gas component was formed on the semiconductor substrate, and the composition inclination layer of the oxygen of the aforementioned metallic oxide of the thickness by which inclination layer thickness is equivalent to five or less atomic layers at the interface was formed. In the manufacture process, a metal membrane is first grown up on a semiconductor substrate at the growth temperature of 250 degrees C or less, oxygen gas is continuously introduced in an epitaxial growth system, and a metallic-oxide film is grown up on the aforementioned metal membrane in oxygen atmosphere. If a metal membrane is thicker than five atomic layer at this time, in the temperature up process after metal membrane membrane formation, it will become easy to diffuse a metal in a semiconductor substrate. Moreover, since oxygen is spread in this metal membrane during membrane formation of the metallic-oxide film after a temperature up, the composition inclination layer of the oxygen of a semiconductor substrate, this metal membrane equivalent to the thickness which is equivalent to five or less atomic layers at the interface of a metallic oxide, and the metallic-oxide film to which oxygen reacted is formed. Therefore, if elements, such as a nonvolatile memory, a capacitor, a thin film superconductivity device, or an optical modulator, are produced using this substrate, the buffer layer which consists of a metallic-oxide film will be thin, and the element of the outstanding property will be obtained.
[0016] and a semiconductor substrate -- a field direction -- or (111) (100) can integrate a nonvolatile memory, a capacitor, a thin film superconductivity device, or an optical modulator by being made of single crystal silicon Moreover, if the metallic-oxide film is made of the oxide and yttrium oxide which have C rare-earth structure, the oxidization dysprosium, or the yttria partially stabilized zirconia, the obtained multilayer semiconductor substrate is suitable as a substrate for element production of a nonvolatile memory, a capacitor, a thin film superconductivity device, or an optical modulator. In addition, as an oxide which has C rare-earth structure, there are oxidization neodium (Nd 2O3), a samarium oxide (Sm 2O3), oxidization europium (Eu 2O3), oxidization pro SHICHIUMU (Pr 2O3), etc. other than a yttrium oxide (Y2 O3) and an oxidization dysprosium (Dy 2O3).
[0017] According to the manufacture method of the multilayer semiconductor substrate concerning this invention, in an epitaxial growth system Pay a semiconductor substrate and a metal raw material and a metal membrane is first grown up on this semiconductor substrate at the growth temperature of 250 degrees C or less. Then, since oxygen gas is introduced in this epitaxial growth system and it was made to grow up a metallic-oxide film on the aforementioned metal membrane in oxygen atmosphere, Since oxidization and **********ing are prevented for the front face of a semiconductor substrate in the process to which the temperature up of the substrate temperature is carried out to the growth temperature (for example, 800 degrees C) of a metallic oxide, a thin and uniform metallic-oxide thin film is formed. Therefore, the outstanding substrate suitable as substrates for production, such as a nonvolatile memory, a capacitor, a thin film superconductivity device, or an optical modulator, is obtained.
[0018] and a field direction -- the suitable substrate for making it integrate with semiconductor devices, such as a nonvolatile memory, a capacitor, a thin film superconductivity device, or an optical modulator, is obtained for or (111) (100) by using a single-crystal-silicon semiconductor substrate Moreover, when it oxidizes as a metal membrane, while growing up the aforementioned metal membrane which consists of a metal used as C rare-earth structure While growing up the film which consists of a metallic oxide of

C rare-earth structure as a metallic-oxide film or growing up the aforementioned metal membrane which consists of an yttrium or a dysprosium as a metal membrane While growing up the film which consists of a yttrium oxide or an oxidization dysprosium as a metallic-oxide film or growing up the aforementioned metal membrane which consists of a zirconium as a metal membrane The outstanding multilayer semiconductor substrate is obtained by growing up the film which consists of a yttria stabilized zirconia as a metallic-oxide film. Furthermore, the multilayer substrate with which it has a thin and uniform metallic-oxide thin film, and the semiconductor substrate is not polluted with the metal atom in a metal membrane is obtained by growing up a metal membrane so that the thickness may turn into thickness which is preferably equivalent to 1.5-2.5 or less atomic layers one to 5 atomic layer. If the thickness of a metal membrane is too as thin as less than one atomic layer, the effect of antioxidizing of a semiconductor substrate front face or etching prevention will not fully be acquired. On the contrary, after metal membrane membrane formation, if the thickness of a metal membrane is too thicker than five atomic layers, when substrate temperature is raised, it will become easy to diffuse the metal atom from a metal membrane to the inside of a semiconductor substrate.

[0019] moreover, by **(ing) the front face of a semiconductor substrate in dilute-acid solutions and FUKKA ammonium water, such as rare fluoric acid, beforehand, or irradiating a hydrogen radical on the front face of a semiconductor substrate, and carrying out hydrogen termination processing on the substrate front face, the effect of antioxidizing of a semiconductor substrate becomes high and the multilayer semiconductor substrate which has the thin and uniform metallic-oxide film which is the thickness about a 1 - pentatomic layer on a pure semiconductor substrate front face is obtained

[0020]

[Example] The place by which gives an example to below and it is characterized [of this invention] is clarified. In addition, this invention cannot be overemphasized by that it is not what is restricted in any way according to each following example.

[0021] (Example 1) After carrying out evacuation of the inside of a deposition chamber and considering as vacuum atmosphere (pressure : 1x10 -6 Pa) first using well-known vacuum evaporationo equipment, Y (yttrium) film was grown up on the semiconductor substrate which a field direction becomes from the single crystal silicon of (111). The simple substance (metal) of Y was used for the raw material. The temperature of a substrate was 250 degrees C. The thickness of grown-up Y film was 4A. In addition, the semiconductor substrate performed wet processing from acids, such as rare fluoric acid (dil HF), beforehand, and what carried out hydrogen termination processing of the front face of a semiconductor substrate was used for it.

[0022] Then, it is O2 in a deposition chamber. It is Y2 O3 on Y film which introduced gas and was previously grown up in oxygen atmosphere (pressure : 1x10 -2 Pa). The film was grown epitaxially. The simple substance of Y was used for the raw material. Substrate temperature was 700 degrees C. Grown-up Y2 O3 Membranous thickness was 200A.

[0023] Drawing having shown typically the result of RHEED (reflective high-speed electron diffraction) in drawing 2 (a) at XRD (X diffraction) and drawing 2 (b) is shown. In the result of XRD, only the peak of Y2 O3 (444) and the peak of Si (111) substrate have appeared, and the clear pattern has appeared in the result of RHEED so that drawing 2 (a) and (b) may show. These results show that the epitaxial film of uniform Y2O3 orientation (111) with few dry areas has grown to be a front face on Si (111) substrate.

[0024] (Example 2) Dy (dysprosium) film was first grown up like the above-mentioned example 1 on the semiconductor substrate (the acid performed wet processing for the front face beforehand, and hydrogen termination processing of the front face of a semiconductor substrate was carried out.) which a field direction becomes from the single crystal silicon of (111) in vacuum atmosphere (pressure : 1x10 -6 Pa). The simple substance (metal) of Dy was used for the raw material. The temperature of a substrate was 250 degrees C. The thickness of grown-up Dy film was 4A.

[0025] Then, it is Dy 2O3 on Dy film in oxygen atmosphere (pressure : 1x10 -2 Pa). The film was grown epitaxially. The simple substance of Dy was used for the raw material. Substrate temperature was 700 degrees C. Grown-up Dy 2O3 Membranous thickness was 200A.

[0026] Drawing having shown the result of RHEED in drawing 3 (a) typically at XRD and drawing 3 (b) is shown. In the result of XRD, only the peak of Dy 2O3 (444) and the peak of Si (111) substrate have appeared, and the clear pattern has appeared in the result of RHEED so that drawing 3 (a) and (b) may show. These results show that the epitaxial film of uniform Dy2 O3 orientation (111) with few dry areas has grown to be a front face on Si (111) substrate.
[0027] (Example 3) Zr film was first grown up like the above-mentioned example 1 on the semiconductor substrate (the acid performed wet processing for the front face beforehand, and hydrogen termination processing of the front face of a semiconductor substrate was carried out.) which a field direction becomes from the single crystal silicon of (100) in vacuum atmosphere (pressure : 1x10 -6 Pa). The simple substance (metal) of Zr was used for the raw material. The temperature of a substrate was 250 degrees C. The thickness of grown-up Zr film was 4A.
[0028] Then, it is O2 in a deposition chamber. Gas was introduced and the YSZ (yttria stabilized zirconia (composition; Y2 O3 : ZrO 2 = 9:91 (mole ratio))) film was grown up on Zr film previously grown up in oxygen atmosphere (pressure : 5x10 -3 Pa). The simple substance of YSZ was used for the raw material. Substrate temperature was 800 degrees C. The thickness of the grown-up YSZ film was 200A.
[0029] Drawing having shown the result of RHEED in drawing 4 (a) typically at XRD and drawing 4 (b) is shown. Only the peak of YSZ (200) has appeared in the result of XRD, and the clear pattern has appeared in the result of RHEED so that drawing 4 (a) and (b) may show (in addition in the result (drawing 4 (a)) of XRD, the peak of Si (100) substrate has not appeared). These results show that the epitaxial film of uniform YSZ (100) orientation with few dry areas has grown to be a front face on Si (100) substrate.
[0030] As mentioned above, in the front face, any sample understands that the uniform epitaxial film with few dry areas is growing about each sample (multilayer semiconductor substrate) obtained in the above-mentioned examples 1-3.
[0031] The multilayer semiconductor substrate which has the property which was excellent as a substrate used in case according to the above-mentioned example the buffer layer which consists of a thin and uniform metallic-oxide thin film can be formed on a semiconductor substrate and a nonvolatile memory, a capacitor, a thin film superconductivity device, or an optical modulator is produced by growing up a metal thin film in vacuum atmosphere on a semiconductor substrate, and growing up a metallic-oxide thin film in oxygen atmosphere on it further is obtained.
[0032] In addition, the metal used as an evaporation source (raw material) is not restricted to the above-mentioned example.
[0033] Moreover, the semiconductor substrate of not restricting to a silicon substrate is natural.
[0034]
[Effect of the Invention] Since the multilayer semiconductor substrate which has a thin and uniform metallic-oxide film on a semiconductor substrate is obtained according to the multilayer semiconductor substrate concerning this invention, elements, such as a nonvolatile memory which was excellent in the property, a capacitor, a thin film superconductivity device, or an optical modulator, are obtained by using this substrate. Moreover, such various elements and a semiconductor device can be integrated.
[0035] According to the manufacture method of the multilayer semiconductor substrate concerning this invention, a thin and uniform metallic-oxide film is formed on a semiconductor substrate, and the outstanding substrate suitable as substrates for production, such as a nonvolatile memory, a capacitor, a thin film superconductivity device, or an optical modulator, is obtained.

[Translation done.]

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PRIOR ART

[Description of the Prior Art] When producing a highly efficient nonvolatile memory, a capacitor or an optical modulator, etc., the thing of the structure which carried out the laminating of the buffer thin film, metal thin film, and ferroelectric thin film which consist of a metallic-oxide film on a GaAs single crystal substrate one by one is known as an usable stacking-tendency ferroelectric thin film as indicated by JP,6-196648,A.

[0003] In producing the element of such structure on the semiconductor substrate which consists of single crystal silicon, it grows a metallic-oxide film epitaxially by the vacuum deposition, the spatter, etc. on a semiconductor substrate first. Conventionally, the sintered compact of a metallic oxide etc. is used for the target of the evaporation source and sputtering. However, when heating the above-mentioned evaporation source and evaporating it with an electron beam etc., or when carrying out the ion bombardment of the target, the maceration of the oxygen from an oxide happened, the front face of a silicon semiconductor substrate will oxidize by the dissociated oxygen, and there was a trouble that a metallic-oxide film with a sufficient property was not obtained.

[0004] After forming 80A metal strontium to this problem on the silicon semiconductor substrate which has an oxide film on a front face, it is forming a strontium-titanate epitaxial film. It is indicated that make the oxide film of a silicon semiconductor substrate front face return with metal strontium, and a good buffer film is obtained (the 13th material science and 109 pages - 114 pages (Proceedings of the 13 th Symposium on Material Science and Engineering) (1994) of engineering symposium collected works). However, by this method, since the thickness of metal strontium is as thick as 80A, a composition gap of a strontium-titanate film will arise. moreover, the thickness of a strontium-titanate film conjectures about 1000A -- having (not clearly written in the paper) -- as a buffer film, it is thick

[0005] Moreover, it is 4A Zr1-x Yx at a room temperature by the sputtering method on a silicon semiconductor substrate. The technology which forms membranes and forms a 2400A yttria partially-stabilized-zirconia epitaxial film on it is indicated (229 Thin Solid Films (Thin Solid Films), 17 pages - 23 pages (1993)). However, since the sputtering method is used for this method, the inert gas concentration in a film becomes high with several %, and a problem produces it on the electrical property when forming a ferroelectric film, a superconductor film, etc. further and element-izing on this, and reliability. Moreover, this technology of the thickness of a yttria partially-stabilized-zirconia film is also thick as 2400A and a buffer film.

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1] O2 It is drawing showing the property of the reaction on gas and the front face of silicon.
[Drawing 2] It is drawing having shown the result of an example 1, and drawing in which drawing 2 (a) shows the result of XRD, and drawing 2 (b) are drawings showing the result of RHEED typically.
[Drawing 3] It is drawing having shown the result of an example 2, and drawing in which drawing 3 (a) shows the result of XRD, and drawing 3 (b) are drawings showing the result of RHEED typically.
[Drawing 4] It is drawing having shown the result of an example 3, and drawing in which drawing 4 (a) shows the result of XRD, and drawing 4 (b) are drawings showing the result of RHEED typically.
[Description of Notations]
I Antioxidizing field
II Oxidization field
III Etching field
IV Oxidization field

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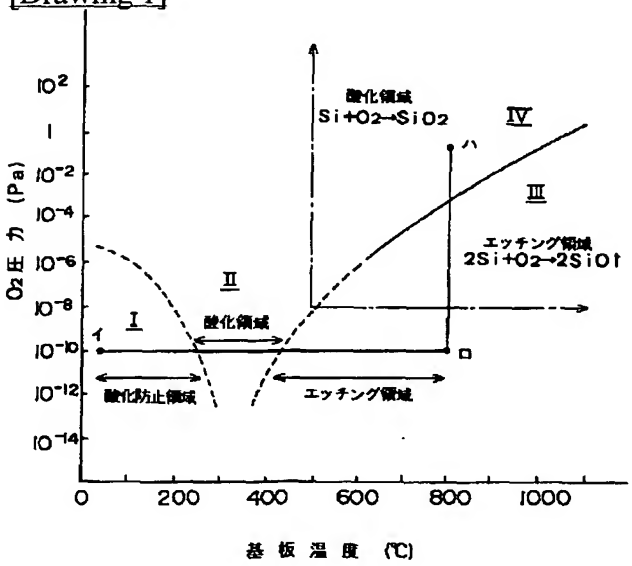
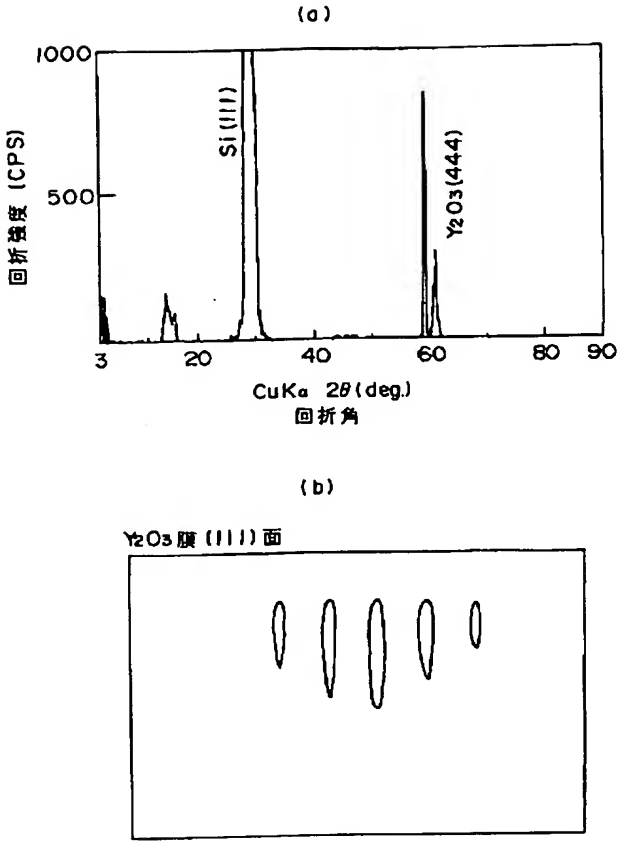
DRAWINGS

[Drawing 1]

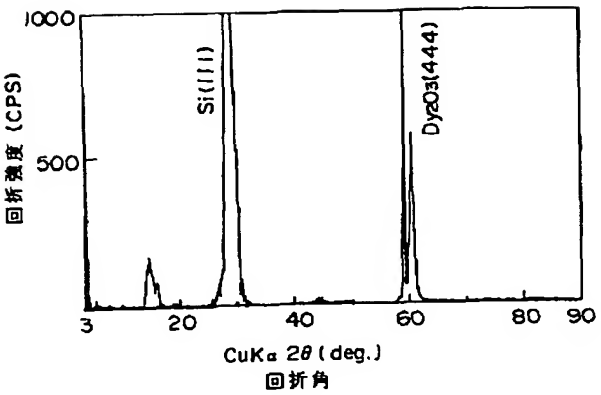


[Drawing 2]


(a)
1000
500
回折強度 (CPS)
Si(111)
Y2O3(444)
3
20
40
60
80
90
CuKα 2θ(deg.)
回折角
(b)
Y2O3膜(111)面

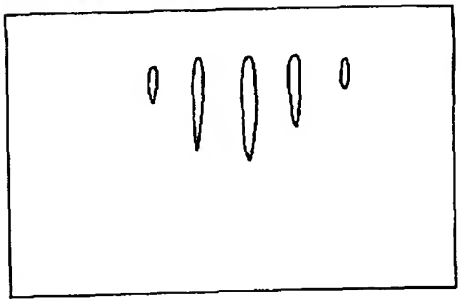
[Drawing 3]

(a)



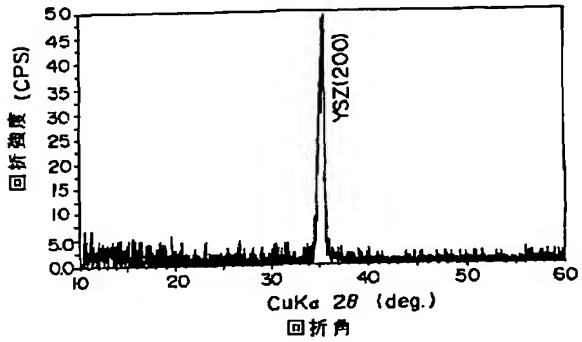
(b)

Dy2O3膜 (111) 面



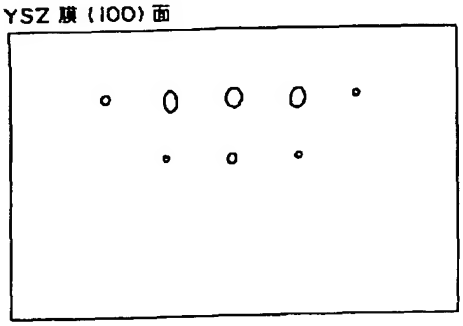
[Drawing 4]

(a)



(b)

YSZ 膜 (100) 面



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